Theory of diamagnetism in an asymmetrical vertical quantum dot molecule

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We consider a system of two vertical lens-shaped quantum dots. The quantum dots have substantially different diameters in contrast to most of the investigated structures. We demonstrate theoretically a possibility to relocate the ground-state electronic wave function from one dot to another by applying an external magnetic field along the structure's growth direction. The relocation correlates with the anticrossing behavior of the two lowest electronic energy states of the quantum dot molecule. This effect originates from a nonuniform diamagnetic shift for the electronic states located in different dots and generates unusual features of the magnetization and magnetic susceptibility of the system. At low temperature, the differential susceptibility of the system has a positive peak. With increasing temperature the peak gradually disappears. From our results it follows the opportunity to design nonmagnetic semiconductor nanostructures with unusual magnetic properties.

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Motivated by the current need for multifunctional nano-based semiconductor devices, an extensive study of components and subsystems to be reconfigurable and controlled dynamically has been performed. In this domain composite materials (metamaterials, artificially structured materials) offer a very promising direction of the future development mainly in the modern optics and information technology sector (see Refs. 1–4 and references therein). Metamaterial properties derive from properties of the constituent materials. Therefore, incorporating into the metamaterials tunable nanosized components (nano-objects) leads to tunable properties of the metamaterials.

Recent advances in semiconductor nanotechnology makes it possible to study in detail different kinds of semiconductor nano-objects and fabricate artificial semiconductor nanostructured metamaterials. Quantum dots and nanorings are nanosized semiconductor structures resembling artificial atoms and magnetic properties of those systems attracted particular attention in recent decades.[5–13] Pairs of high-quality coupled semiconductor quantum dots [quantum dot molecules (QDMs)] were proposed recently to manipulate electronic states by variation in the distance between the dots (*static coherent tuning*)[14–17] or by variation in the external fields[18] (*dynamic coherent tuning*). A vast amount of literature discusses the magnetic phenomena in terms of spin magnetism in quantum dots (see, for instance, Ref. 8 and references therein). However, under certain conditions the diamagnetism can be even more important and very unusual in semiconductor nano-objects. For instance, semiconductor nanorings are *doubly* (*nonsimply*) *connected objects*. When the external magnetic field **B** is in the system growth direction, the center hole enables oscillations of the magnetization (the Aharonov-Bohm effect[19]) and a positive peak in the differential magnetic susceptibility (DMS) of the single electron nanorings.[11–13,20]

Here we show that DMS of a single electron vertical QDM also can demonstrate the positive peak if the incorporated dots have substantially different radial dimensions and the external magnetic field **B** is in the system growth direction. The important difference from nanorings is that the molecule is a *totally separated geometrical object* [see the inset of Fig. 1(a)].

To determine the magnetic moment for an isolated molecule, the standard approach is to calculate the total electronic energy of the molecule in the presence of the external magnetic field. The magnetization (total magnetic moment) M and differential magnetic susceptibility χ of the molecule are[21]

$$M = \sum_p \left(-\frac{\partial E_p}{\partial B} \right) f(E_p - \mu) \tag{1}$$

and

$$\chi = \frac{\partial M}{\partial B}, \tag{2}$$

where p stands for the electronic state of the QDM with corresponding energy E_p, $f(E)$ is the Fermi distribution function, and μ is the chemical potential of the system determined from the number of the electrons confined in the QDM by the following equation:

$$N = \sum_p f(E_p - \mu). \tag{3}$$

In this study we consider a system of two circular vertical lens-shaped quantum dots. The calculations are done for the three-dimensional model of semiconductor QDM with the hard-wall confinement potential. This allows us to simulate the magnetic properties of the electron energy states in the system of a very asymmetrical shape: the quantum dots have substantially different diameters and different heights in contrast to most of the known simulations. To demonstrate the unusual DMS of the single electron vertical QDM we adopt the effective one-electronic-band effective Hamiltonian,[11,22]

$$\hat{H} = \frac{1}{2}\mathbf{\Pi}_\mathbf{r}\frac{1}{m(E,\mathbf{r})}\mathbf{\Pi}_\mathbf{r} + V(\mathbf{r}) + \frac{\mu_B}{2}g(E,\mathbf{r})\sigma\cdot\mathbf{B}, \tag{4}$$

where $\mathbf{\Pi}_r = -ih\nabla_r + e\mathbf{A}(\mathbf{r})$ is the electron momentum operator, with ∇_r the spatial gradient and $\mathbf{A}(\mathbf{r})$ the vector potential of

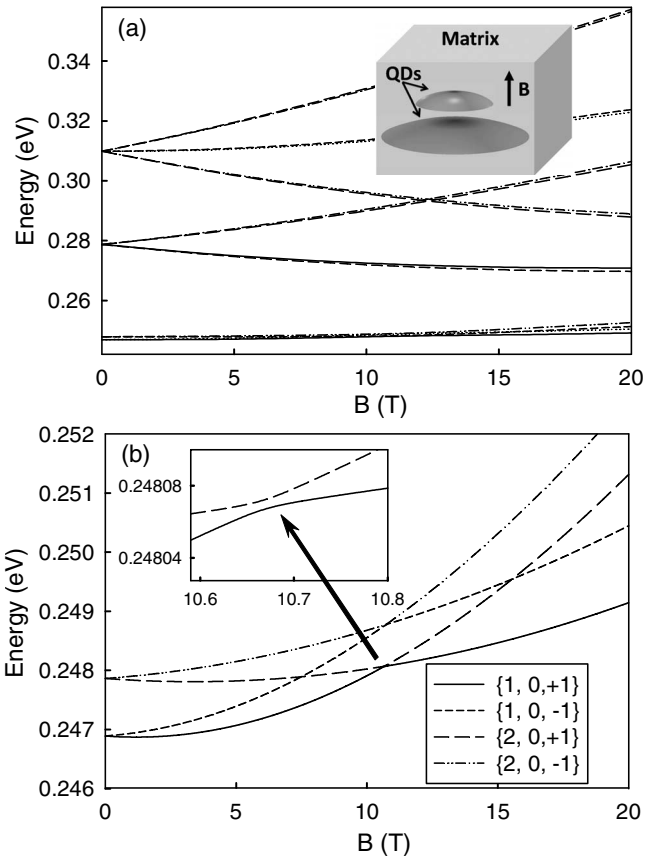

FIG. 1. Electron energy states of asymmetrical QDM as functions on the external magnetic field: (a) 14 lowest states, in inset asymmetrical InAs/GaAs QDM in the external magnetic field **B**; (b) 4 lowest states with $l=0$, in inset region of anticrossing for $\{1,0,+1\}$ and $\{2,0,+1\}$ states.

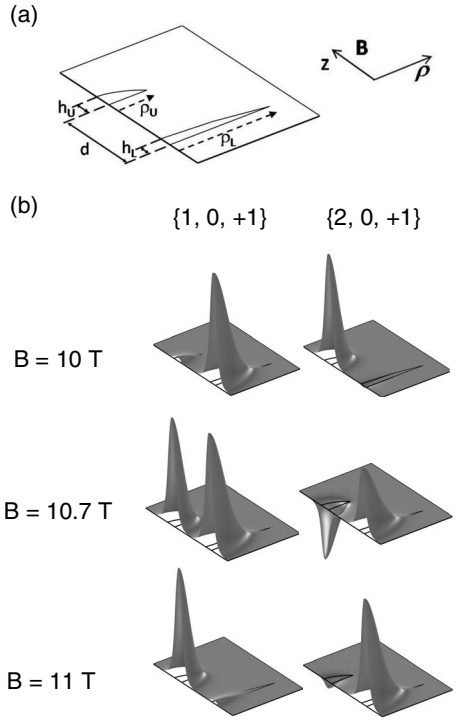

FIG. 2. Electronic wave functions in the asymmetrical QDM: (a) QDM projection onto (ρ,z) plane; (b) transformation of the lowest electron energy state wave functions near the anticrossing point B_C.

the magnetic field $\mathbf{B}=\mathrm{curl}\,\mathbf{A}$, $m(E,\mathbf{r})$ and $g(E,\mathbf{r})$ are the electron energy and position dependent electron effective mass and the Landé factor, $V(\mathbf{r})$ is the confinement potential, and σ is the vector of the Pauli matrixes. The Hamiltonian (with certain modifications) enables us to describe the lowest electronic energy states in different *III-V semiconductor* nano-objects.[11,12,23] As it follows from our results below this relatively simple approach allows us to concentrate upon the most important peculiarities of the QDM magnetic properties and discover some *principal conditions* for the DMS's unusual behavior, which is the main goal of this study. Further details can be found in Ref. 11.

In our model the magnetic field is directed along the system z axis, and we can treat the problem in cylindrical coordinates (ρ,z,ϕ). The wave function can be represented as

$$\Psi(\mathbf{r}) = \Phi(\rho,z)\exp(il\phi), \tag{5}$$

where $l=0,\pm1,\pm2,\ldots$ is the orbital quantum number. This leads to a two-dimensional computational problem in the (ρ,z) coordinates [see Fig. 2(a)] and forms a set of the quantum numbers $p=\{n,l,s\}$, where n is the principal quantum number and $s=\pm1$ refers to the orientation of the electron spin along the z axis. Dimensions of the dots we choose are close to known experimental structures.[24] The most important difference of this molecule from those usually discussed in the literature is that our quantum dots have different radii $\rho_L>\rho_U$ and heights $h_L<h_U$ [L and U stand for the "lower" and "upper" dots in Fig. 2(a)]. So the system is asymmetrical in z direction. Here we present our results including the lowest electron energy states of a system assembled from the lens-shaped dots with $\rho_L=25$ nm, $\rho_U=9.5$ nm, $h_L=3$ nm, $h_U=4$ nm, and different interdot (base-to-base) distance d.

To demonstrate the feasibility of the unusual diamagnetic response in the asymmetrical QDM we use realistic semiconductor material parameters for InAs/GaAs QDM (as an example); for instance, the band offset of the InAs/GaAs strained heterostructure, corrected to the strain condition band parameters, etc. For the strained InAs inside the dots according to the corrections done in Ref. 25 we choose energy gap $E_{g\mathrm{InAs}}=0.842$ eV, $\Delta_{\mathrm{InAs}}=0.39$ eV, and $m_{\mathrm{InAs}}(E=0)=0.044m_0$ (E_g and Δ are the material energy gap and the spin-orbit split-off energy and m_0 is the free-electron mass). For the GaAs matrix we take from[26] $E_{g\mathrm{GaAs}}=1.519$ eV, $\Delta_{\mathrm{GaAs}}=0.341$ eV, and $m_{\mathrm{GaAs}}(E=0)=0.067m_0$, and the system band offset is $V_0=0.474$ eV. The energy states and wave functions of the electrons confined in the quantum dot molecule are found by the nonlinear iterative method[23,27] using the COMSOL MULTIPHYSICS package.[28]

The asymmetry in the QDM geometry generates interesting features of the lowest energy states with $l=0$, while the upper states ($l\neq0$) demonstrate traditional dependencies on the magnetic field (Fig. 1). For the reason of clarity we consider now only two lowest energy states with the angular momentum $l=0$: $p_1=\{1,0,+1\}$ and $p_2=\{2,0,+1\}$ [see Fig.

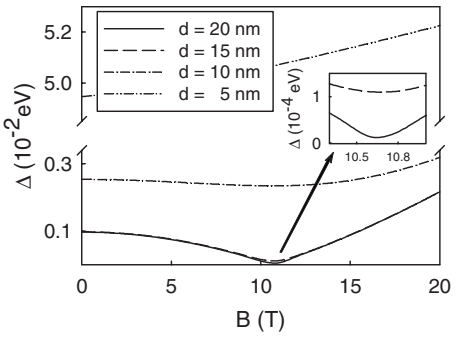

FIG. 3. Energy gap between the lowest energy levels for different interdot distances.

1(b)]. The most important (for the unusual magnetic properties of the QDM) result is the anticrossing between those two states (states of the same symmetry) at $B_C \approx 10.7$ T. The inset of Fig. 1(b) shows a portion of the spectrum near the anticrossing point. This result can be readily understood on the base of the traditional approach to the diamagnetic shift theory in nanosized three-dimensional systems.[21,22,29] In the first approximation of the perturbation theory the diamagnetic shifts of the states with $l=0$ can be presented as $\delta E_p(B) \sim \langle p|\rho^2|p\rangle B^2$, where $\langle\|\rangle$ denotes the quantity being averaged with the state wave function at zero magnetic field. It can be seen from Fig. 2(b) that the state p_1 at $B<B_C$ corresponds to the wave function located in the lower dot. At the same time, the state p_2 at $B<B_C$ corresponds to the wave function located in the upper dot. Clearly, for our QDM, when $E_{p_1}(B=0)<E_{p_2}(B=0)$ and $\langle p_1|\rho^2|p_1\rangle > \langle p_2|\rho^2|p_2\rangle$, those two levels should converge when the magnetic field is increasing. Having the same symmetry according to the general theory[21] the levels anticross at certain B_C. When $B \simeq B_C$ the wave function of the state p_1 relocates from the lower dot [Fig. 2(b)] to the upper one. The opposite relocation can be traced for the second state p_2: from the upper dot to the lower one. So the magnetic field acts as a *dynamic coupling factor* for energy states localized in different dots on the analogy of the interdot distance in the static approach.[16]

Clearly, the anticrossing and relocation can be achieved only when the lateral dimensions of the dots are substantially different. At the same time the energy gap between two states $\Delta(B)=E_{p_2}-E_{p_1}$ at zero magnetic field can be tuned by an adjustment in the dots' heights and distance between them. Figure 3 shows the gap dependence on the external magnetic field for different interdot distances. Obviously, the unusual anticrossing requires some separation of the dots. For the distance $d \lesssim 10$ nm the wave-function relocation disappears and no actual anticrossing can be achieved by change in the magnetic field. So the effect has inherently three-dimensional nature and can be predicted and explained only within the proper three-dimensional description.

The anticrossing of the lowest energy levels in the system with one electron generates very unusual features of the total magnetic moment (1) near the anticrossing point (see Fig. 4). At low temperature, the magnetic moment demonstrates a jump which is similar to that theoretically predicted[11,20] and experimentally observed[13] in single electron nanorings.

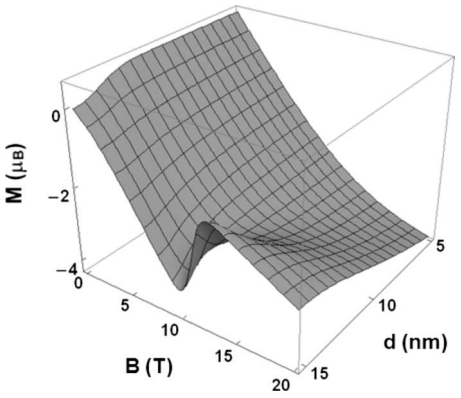

FIG. 4. Magnetic moment of single electron asymmetrical quantum dot molecule as a function on the magnetic field and distance between dots at 0.5 K.

When temperature increases the jump gradually disappears and the system returns to the conventional diamagnetic behavior. The magnitude of the jump can reach up to few μ_B per QDM, which is within the range of present day detectability.[13] It should be noted that the electronic spin contribution is naturally included in our consideration of the total magnetic moment [see Hamiltonian (4)].

Finally we consider the important issue of the unusual diamagnetism of the asymmetrical QDM. The DMS is remarkably nonlinear. The above-described jump in the magnetization of the single electron QDM generates a peak in DMS (2). At zero temperature, the calculated DMS has a deltalike "paramagnetic" peak at B_C. With increasing temperature the peak remains Lorentz-type shaped and gradually disappears. In Fig. 5 we present the DMS at different temperatures. Most importantly, the peak can reach the *positive* value at low temperatures. Calculations showed that for our QDM at the temperature of 0.5 K the magnitude of the peak is about $+9\times10^{-5}$ eV T^{-2}.

We stress that the unusual diamagnetic properties of the single electron QDM follow from the system geometry and general quantum physics. Obviously, we can expect the effect in any nano-object (*not* only in III-V semiconductor QDMs) where the lowest energy states of the same cylindrical symmetry are close and the nonuniform diamagnetic shift

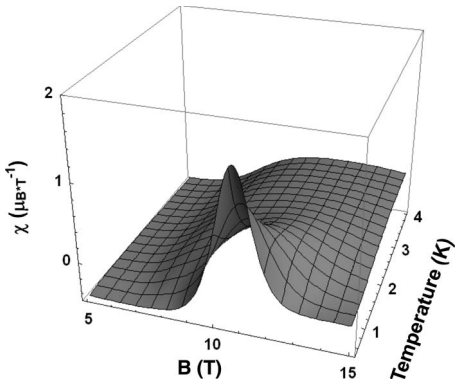

FIG. 5. Magnetic susceptibility of single electron asymmetrical quantum dot molecule as a function on magnetic field and temperature ($d=20$ nm).

(the wave-function relocation driven by magnetic field) is possible. Those are two main conditions of the effect appearance. In general, the approximate objects' shapes and sizes (as well as the material parameters) always have deviations. However, the present day technology for InAs/GaAs quantum dot molecules can provide us with a wide range of opportunities to tune the system geometry and parameters. This makes InAs/GaAs quantum dot molecules the most probable candidates for the experimental observation of the effect.

In conclusion, our calculation results demonstrate that in a three-dimensional asymmetrical quantum dot molecule the nonuniform diamagnetic shift of the lowest energy levels in different dots leads to their anticrossing which produces the positive peak of the differential magnetic susceptibility. Specifically, III-V semiconductor quantum dot molecule systems can be designed to observe this effect. The geometry is crucial for such investigations and should be properly tuned and controlled. It follows from this theoretical study that experimental investigation of the magnetic properties of asymmetrical semiconductor quantum dot molecules will yield interesting results and can be useful for further fabrication of materials with unusual magnetic properties.

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